# Montmorillonite K-10 clay as reusable heterogeneous catalyst for the microwavemediated solventless synthesis of phthalazinetetraones

### Davood Habibi, Nosratollah Mahmoodi, and Omid Marvi

**Abstract:** Different phthalazino[2,3-b]phthalazine-5,7,12,14-tetraones were synthesized in a simple and environmentally benign method from the reaction of some phthalic anhydrides with semicarbazide or thiosemicarbazide using montmorillonite K-10 clay as solid heterogeneous acidic catalyst and microwaves under solvent-free conditions in good yields and short reaction times. The present method has many obvious advantages compared with those reported in the literature, including high efficiency, higher yield, operational simplicity, environmental benignity, and the possibility of recycling the solid clay. The solid clay catalyst used in the first cycle of the reactions was successfully recovered and reused in the second cycle, showing a gradual decrease in activity.

*Key words:* montmorillonite K-10 clay, microwave, solvent-free condition, phthalazinetetraones, semicarbazide, thiosemicarbazide.

**Résumé :** Faisant appel à une méthode simple et environnementalement bénigne, on a réalisé la synthèse de divers phtalazino[2,3-b]phtalazine-5,7,12,14-tétrones avec de bons rendements en faisant réagir des anhydrides phtaliques avec de la semicarbazide ou de la thiosemicarbazide, en présence d'argile de montmorillonite K-10 comme catalyseur acide solide et hétérogène, dans des conditions sans solvant, sous l'influence de microondes et des temps de réaction courts. Cette méthode présente des avantages évidents par comparaison avec celles rapportées dans la littérature, dont le fait qu'elle est environnementalement bénigne, sa grande efficacité, son rendement plus élevé, sa simplicité d'opération et la possibilité de recycler l'argile solide. De plus, de l'argile utilisée dans un premier cycle de réactions a été réutilisée avec succès dans un second cycle, ce qui démontre que la diminution de son activité n'est que graduelle.

*Mots clés* : argile de montmorillonite K-10, microonde, condition sans solvant, phtalazinetétrone, semicarbazide, thiosemicarbazide.

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### Introduction

In the past few decades, the synthesis of new heterocyclic compounds has been a subject of great interest because of their wide applicability. Heterocyclic compounds occur very widely in nature and are essential to life. Nitrogencontaining heterocyclic molecules constitute the largest portion of chemical entities and are part of many natural products, fine chemicals, and biologically active pharmaceuticals that are vital for enhancing quality of life (1).

Heterocyclic skeletons provide scaffolds on which pharmacophores can be arranged to yield potent and selective drugs (2). In this regard, the phthalazine scaffold has shown its potential as a useful structure for the generation of

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druglike libraries in the drug discovery process (3). Moreover, heterocyclic fused phthalazines have been found effective in the inhibition of p38 MAP kinase (4), the selective binding of the GABA receptor (5), as antianxiety drugs (6), as antitumor agents (7), and as high-affinity ligands to the  $\alpha_{2}\delta$ -1 subunit of the calcium channel (8). Despite their wide applicability among the various reported studies on fused nitrogen heterocycles (9), only a limited number of phthalazinetetraone derivatives have been synthesized (10). Cardia et al. (11) presented the most recent method for the synthesis of phthalazino[2,3-b]phthalazine-5,7,12,14-tetraone. Most of these procedures involve the use of corrosive reagents such as polyphosphoric and acetic acids and the reported yields are far from satisfactory. Therefore, the use of inexpensive and environmentally safe solid acids would extend the scope of these transformations. Recently, the use of solid acidic catalysts like clays, zeolites, and ion-exchange resins, has received much research interest in different areas of organic synthesis because of their environmental compatibility, reusability, greater selectivity, noncorrosiveness, low cost, and ease of handling.

In particular, the clay catalysts make the reaction process more convenient, economical, environmentally benign, and





act as both Bronsted and Lewis acids in their natural and ion-exchanged forms, enabling them to function as efficient catalysts for various transformations (12). Montmorillonite clays have been used as catalysts for a number of organic reactions and offer several advantages over classical acids: strong acidity, noncorrosive properties, recyclability, low cost, mild reaction conditions, high yields and selectivity, and the ease of setup and workup (13). Neat reactions, on the other hand, have their own practical problems such as overheating and charring of compounds because of ineffective dissipation of energy.

Also, in the last few years a growing interest has been shown in the use of microwave irradiation in organic synthesis (14). Microwave-assisted rapid organic reactions constitute an emerging technology that makes experimentally and industrially important organic syntheses more effective and more ecofriendly than conventional reactions (15). In addition, microwave-mediated solvent-free synthesis offers advantages for reducing hazardous explosions and the need to to remove solvents with high boiling points from the reaction mixtures. Moreover, coupling solventless synthesis with microwaves shows benefits of shorter reaction times, uniform heating, and higher yields (16, 17).

In continuation of our study on microwave-assisted reaction on solid surfaces (18, 19), herein we describe a convenient and simple method to synthesize some of the phthalazino[2,3-*b*]phthalazine-5,7,12,14-tetraones using montmorillonite clay K-10 as a heterogeneous acidic catalyst and microwave irradiation under solvent-free conditions. The solid montmorillonite clay was successfully recovered later and reused in subsequent reactions.

### **Results and discussion**

Different substituted phthalic anhydrides and either semicarbazide or thiosemicarbazide reacted for 3-7 min under solvent-free conditions using microwave irradiation at 180 °C on montmorillonite K-10 clay (Scheme 1). Under classical heating conditions, the reactions were carried out at a similar temperature but for longer reaction times (40–70 min) and lower yields.

The polarity of the carbonyl group of semicarbazide is greater than its thiocarbonyl analog in thiosemicarbazide, so the  $-NH_2$  group in thiosemicarbazide is a better nucleophile

than the -NH<sub>2</sub> group in semicarbazide and hence will produce better yields. It seems that the nature of the substituents of phthalic anhydride has some effects on this reaction. It is of interest to note that the presence of electron-withdrawing groups like Cl and NO<sub>2</sub> gave high product yields and short reaction times compared with the unsubstituted phthalic anhydride. The use of the solid clay as an efficient heterogeneous acidic solid catalyst as well as microwave irradiation offer high product yields compared with conventional procedures. A comparison has been given in the general procedure section. Among the other advantages of our method is the recyclability of solid clay. The clay catalyst used in the first cycle was recovered by filtration, washed with methanol, and reused. Recyclability details of the clay have been explained in the footnote of Table 1. It is notable that all attempts to prepare the products in the absence of montmorillonite K-10 clay failed (despite 15 min irradiation at 180 °C). Therefore, the role of montmorillonite K-10 clay as a solid catalyst in this reaction is essential. Furthermore, the use of reusable solid acid makes this method quite simple, more convenient, and economically viable. Finally, in a second series of experiments the reactions were carried out under the conventional heating conditions using an oil bath at similar temperatures (170-180 °C, solvent-free) without microwave irradiation. It was observed that under classical heating the reactions gave the products with relatively lower yields and longer reaction times. Moreover, the reactions required continuous stirring to prevent charring. Comparison of the results clearly shows the preference of microwave irradiation over conventional heating. The results for the obtained products under both conditions are listed in Table 1.

### Experimental

Melting points were measured on an Electrothermal 9100 apparatus. The IR spectra were obtained on a PerkinElmer FT IR GX instrument using KBr discs. The <sup>1</sup>H NMR and <sup>13</sup>C NMR spectra were recorded on a FT-NMR JEOL FX 90Q spectrometer using TMS as internal standard ( $\delta$  given in ppm). Mass spectra were measured with a GCMS Agilent 5973N instrument. Elemental analyses were performed using a Heraeus CHN Rapid analyzer. Chemicals were purchased from the Sigma-Aldrich and Merck chemical companies and used without further purification. Montmorillonite K-10 clay

Table 1	1.	The	yields	and	reaction	times	of	compounds	a-	j
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					Yield (%) <sup>a</sup>	
No.	Anhydride	Reagent	Compound <sup>b</sup>	Time (min)	Fresh clay	Recovered clay
1	Phthalic anhydride	Semicarbazide	а	7.0 (70)	61 (43)	54 (32)
2	Tetrachloro-phthalic anhydride	Semicarbazide	b	3.5 (45)	73 (56)	64 (38)
3	3-Nitro-phthalic anhydride	Semicarbazide	c	5.5 (60)	69 (52)	60 (35)
4	4-Nitro-phthalic anhydride	Semicarbazide	d	5.0 (55)	67 (48)	57 (34)
5	3-Fluoro-phthalic anhydride	Semicarbazide	e	6.0 (65)	65 (45)	58 (30)
6	Phthalic anhydride	Thiosemicabazide	f	5.0 (55)	67 (48)	61 (35)
7	Tetrachloro-phthalic anhydride	Thiosemicabazide	g	3.0 (40)	82 (61)	77 (43)
8	3-Nitro-phthalic anhydride	Thiosemicabazide	h	4.0 (50)	74 (53)	58 (34)
9	4-Nitro-phthalic anhydride	Thiosemicabazide	i	3.5 (45)	73 (55)	56 (39)
10	3-Fluoro-phthalic anhydride	Thiosemicabazide	j	4.0 (50)	70 (54)	64 (36)

Note: The results given in parentheses correspond to those reactions that were carried out under classical heating conditions (solvent-free, 170–180 °C, oil bath, without microwave irradiation).

"Isolated yields after recrystallization. The solid clay portion used in the first cycle was filtered off, washed with methanol ( $2 \times 20$  mL) under stirring for 1 h, and dried at 120 °C for 5 h under reduced pressure, to be reused in the subsequent reaction, which showed a gradual decrease in activity.

 $^{b}(\mathbf{a}, \mathbf{f})$  Phthalazino[2,3-*b*]phthalazine-5,7,12,14-tetraone; (**b**, **g**) octachlorophthalazino[2,3-*b*]phthalazine-5,7,12,14-tetraone; (**c**, **h**) 1,11-dinitro-

phthalazino[2,3-*b*]phthalazine-5,7,12,14-tetraone; (**d**, **i**) 2,10-dinitro-phthalazino[2,3-*b*]phthalazine-5,7,12,14-tetraone; (**e**, **j**) 1,11-difluorophthalazino[2,3-*b*]phthalazine-5,7,12,14-tetraone; (**e**, **j**) 1,11-difluorophthalazine[2,3-*b*]phthalazine-5,7,12,14-tetraone; (**e**, **j**) 1,11-difluorophthalazine[2,3-*b*]phthalazine[2,3-*b*]phthalazine[2,3-*b*]phthalazine[2,3-*b*]phthalazine[2,3-*b*]phthalazine[2,3-*b*]phthalazine[2,3-*b*]phthalazine[2,3-*b*]phthalazine[2,3-*b*]phthalazine[2,3-*b*]phthalazine[2,3-*b*]ph

was purchased from Fluka chemical company [surface area: 200 m<sup>2</sup>/g; pH 2.5–3.5; chemical composition (average value): SiO<sub>2</sub> (73.0%), Al<sub>2</sub>O<sub>3</sub> (14.0%), Fe<sub>2</sub>O<sub>3</sub> (2.7%), CaO (0.2%), MgO (1.1%), Na<sub>2</sub>O (0.6%), K<sub>2</sub>O (1.9%)].

## Typical procedure for microwave-mediated synthesis of phthalazino[2,3-b]phthalazine-5,7,12,14- tetraones

In a typical experiment, phthalic anhydride (2.2 mmol, 0.330 g), thiosemicarbazide (1 mmol, 0.091 g), and montmorillonite K-10 clay (1 g) were mixed in a mortar and the mixture was placed in a pyrex tube and introduced into a Synthewave 402<sup>®</sup> (Prolabo, France) single mode focused microwave reactor for 5 min at 180 °C (monitored temperature (20)) with continuous rotation. The reaction mixture was then allowed to cool to room temperature. The resulting product was extracted into dichloromethane  $(2 \times 25 \text{ mL})$ , filtered off, and the solvent was removed by rotary evaporation. The obtained product, phthalazino[2,3-b]phthalazine-5,7,12,14-tetraone (f), was very thoroughly washed with distilled water, dried in an oven, and recrystallized from acetic acid. Melting point 332-336 °C. Yield with the fresh K-10 clay, 67% and 0.195 g; yield with the recovered K-10 clay, 61% and 0.178 g (lit. values (11) mp 343 to 344 °C, yield 55%, reaction time 1.5 h using polyphosphoric acid). The solid clay portion was washed with methanol and dried at 120 °C under reduced pressure to be reused in the subsequent reactions, which showed a gradual decrease in activity (Table 1). With this procedure compounds, **a**-**j** were synthesized. The physical data (mp, IR, NMR, and MS) of known compounds **a** and **f** were found to be identical with those of authentic samples. Unknown compounds were characterized by mp, spectra (IR, NMR, and MS), and elemental analyses.

#### Phthalazino[2,3-b]phthalazine-5,7,12,14-tetraone (a, f)

Melting point  $3\overline{32}$ -336 °C. IR (KBr, cm<sup>-1</sup>) v: 3092.7, 1853.5, 1792.6, 1763.5, 1686.4, 1597.4, 1471.3, 1258.7, 1109.8, 907.2. <sup>1</sup>H NMR (DMSO- $d_6$ )  $\delta$ : 7.81–7.98 (8H, m, Ar). <sup>13</sup>C NMR (DMSO- $d_6$ )  $\delta$ : 123.0, 129.3, 134.8, 168.9.

MS m/z: 292 (M<sup>+</sup>). Anal. calcd. for C<sub>16</sub>H<sub>8</sub>N<sub>2</sub>O<sub>4</sub>: C 65.76, H 2.76, N 9.58; found: C 65.12, H 2.60, N 9.48.

## Octachlorophthalazino[2,3-b]phthalazine-5,7,12,14-tetraone (b, g)

Melting point 317–321 °C. IR (KBr, cm<sup>-1</sup>) v: 1846.7, 1835.4, 1775.7, 1757.76, 1373.9, 1299.3, 1233.8, 923.3. <sup>13</sup>C NMR (DMSO- $d_6$ )  $\delta$ : 170.24, 139.92, 135.46, 127.41. MS m/z: 568 (M<sup>+</sup>). Anal. calcd. for C<sub>16</sub>Cl<sub>8</sub>N<sub>2</sub>O<sub>4</sub>: C 33.80, N 4.92; found: C 33.67, N 4.53.

## 1,11-Dinitrophthalazino[2,3-b]phthalazine-5,7,12,14-tetraone (c, h)

Melting point 271–274 °C. IR (KBr, cm<sup>-1</sup>): 3172.9, 3026.24, 2922.0, 1669.9, 1603.3, 1578.1, 1560.8, 1491.6, 1336.5, 1298.2, 825.6. <sup>1</sup>H NMR (DMSO- $d_6$ )  $\delta$ : 7.92–8.63 (6H, m, Ar). <sup>13</sup>C NMR (DMSO- $d_6$ )  $\delta$ : 167.71, 167.43, 148.45, 136.85, 133.39, 132.70, 132.52, 129.61. MS *m*/*z*: 382 (M<sup>+</sup>). Anal. calcd. for C<sub>16</sub>H<sub>6</sub>N<sub>4</sub>O<sub>8</sub>: C 50.26, H 1.57, N 14.56; found: C 50.14, H 1.38, N 14.33.

# 2,10-Dinitrophthalazino[2,3-b]phthalazine-5,7,12,14-tetraone (d, i)

Melting point 263–267 °C. IR (KBr, cm<sup>-1</sup>) v: 3106.3, 3006.5, 1641.8, 1626.7, 1598.7, 1539.9, 1466.6, 1355.1, 1188.4, 1061.3, 791.9. <sup>1</sup>H NMR (DMSO- $d_6$ )  $\delta$ : 8.36–8.94 (6H, m, Ar). <sup>13</sup>C NMR (DMSO- $d_6$ )  $\delta$ : 162.52, 162.43, 154.81, 137.78, 134.66, 133.19, 129.26, 122.34. MS m/z: 382 (M<sup>+</sup>). Anal. calcd. for C<sub>16</sub>H<sub>6</sub>N<sub>4</sub>O<sub>8</sub>: C 50.26, H 1.57, N 14.56; found: C 50.09, H 1.29, N 14.25.

#### 1,11-Difluorophthalazino[2,3-b]phthalazine-5,7,12,14tetraone (e, j)

Melting point 298–302 °C. IR (KBr, cm<sup>-1</sup>): 3021.9, 2921.0, 1671.2, 1603.1, 1577.8, 1560.2, 1491.3, 1336.6, 1299.7, 825.8. <sup>1</sup>H NMR (DMSO- $d_6$ )  $\delta$ : 7.82–8.27 (6H, m, Ar). <sup>13</sup>C NMR (DMSO- $d_6$ )  $\delta$ : 175.27, 163.66, 163.32, 163.08, 136.42, 136.27, 130.52. MS m/z: 328 (M<sup>+</sup>). Anal. calcd. for

 $C_{16}H_6 F_2N_2O_4$ : C 58.53, H 1.82, N 8.53; found: C 58.34, H 1.69, N 8.21.

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